

Cascade of phase transitions induced by magnetic field in graphite

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A strong magnetic field applied perpendicular to the conducting layers of graphite induces an order believed to be a Charge Density Wave (CDW). Previous studies found that the ordered state is restricted to a field window between 25 T and 53 T. By extending magnetoresistance measurements for the first time up to 80 T, we find that this picture is to be substantially revised. Two successive phase transitions, consisting of two distinct ordered states each restricted to a finite field window, are induced by magnetic field. In both states, an energy gap opens up in the out-of-plane conductivity and coexists with an unexpected in-plane metallicity. Such a dichotomy in charge transport contrasts with what is documented in the case of conventional CDW systems. We argue that it may arise as a consequence of edge-state transport in presence of a bulk gap.

Original electronic properties have been reported in stacks of graphene layers. For example, bilayer and trilayer graphene host particular types of quantum Hall effect [1, 2]. When the number of layers is more than eleven[3], the system recovers the properties of bulk graphite, a compensated semi-metal with a tiny three-dimensional Fermi surface and described by the Slonczewski-Weiss-McClure (SWM) band model [4, 5]. In such a macroscopic stack of graphene layers, a perpendicular magnetic field exceeding 7.5 T confines electrons and holes to their lowest Landau levels (LLs). Beyond this so-called quantum limit[6], the electronic spectrum becomes analogue to a one-dimensional system with a variety of possible instabilities[7–9].

In the early eighties, a phase transition in this system was discovered by Tanuma and co-workers who reported a sharp increase in the in-plane magnetoresistance of graphite at $B \approx 25$ T [10]. Numerous experimental studies followed [11–17] and confirmed the existence of a field-induced many-body state beyond a temperature-dependent critical magnetic field. Soon after the initial experimental discovery, Yoshioka and Fukuyama (YF)[18] ascribed this instability to a charge-density-wave (CDW). Such an instability is favored by one-dimensionality, because of the availability of a suitable ($2k_F$) nesting vector. In the original YF scenario, the CDW in adjacent valleys are out of phase, creating a valley density wave state [19], in order to minimize Coulomb energy. In 1998, by further increasing the magnetic field, Yaguchi and Singleton found that the field-induced state is eventually destroyed beyond 53 T[15]. This destruction was attributed to the depopulation of a Landau sub-level within the framework of YF scenario. Possible multiplicity of induced phases and their signa-

tures in the in-plane and out of-plane charge transport remained open questions (for a review see [20]).

In this letter, we extend the previous measurements up to a magnetic field as strong as 80 T and focus on the contrast between in-plane and out-of-plane resistivity. Our results lead to a significant revision of the experimental picture. As the magnetic field is swept, an electronic state is indeed first induced and then destroyed by magnetic field. But this state is immediately followed by another and almost identical one, which is also first induced and then destroyed by the increasing magnetic field. We resolve an activated behavior in the temperature dependence of the out-of-plane conductivity in both of these ordered states, which allows us to quantify the magnitude of the out-of-plane charge gap for the first time. Interestingly, such a gap is absent in the in-plane transport. We argue that this may be the first observation of the three-dimensional counterpart to the two-dimensional edge states known to occur in quantum Hall systems.

Standard four-probe resistivity measurements were performed on both kish and HOPG samples in pulsed magnetic fields at LNCMI-Toulouse. A detailed description of experimental procedure is given in Supplementary Information (S.I.) section A. Fig.1 presents the data for in-plane and out-of plane magnetoresistance up to 70 T for different temperatures. As seen in Fig.1.a, as a consequence of high mobility, the in-plane magnetoresistance is very large. It saturates around $B \sim 20$ T before becoming slightly negative. The onset of the transition to the field-induced state is marked by a sudden increase in resistance. This jump is followed by a drop corresponding to reentry to a low-resistive state at 53 T. Above 53 T, the magnetoresistivity continues to decrease smoothly without indicating any new field scale. These results are in good agreement with the previous data up to 54 T reported by Yaguchi and Singleton[15]

Magnetoresistance along c -axis, R_c , presented in

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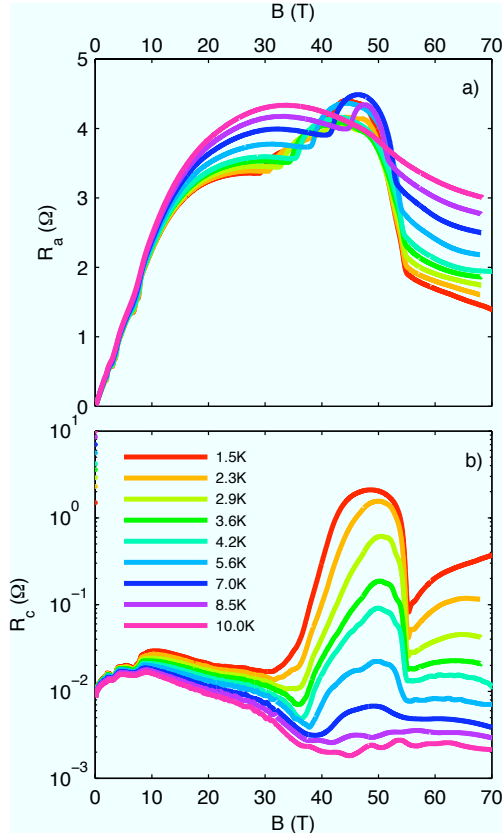


FIG. 1: a) In-plane magnetoresistance of kish graphite (sample K_1) for different temperatures. b) Out-of-plane magnetoresistance resistance of sample (K_2) for various temperature. The magnetic field is oriented along the c -axis. Note the semi-logarithmic scale of the lower panel.

Fig.1.b, shows several remarkable differences with R_a . First, below the onset of transition, the out-of-plane magnetoresistance is much smaller than the in plane magnetoresistance. This is not surprising, since the Lorentz force does not affect the electron motion along the magnetic field[23]. See the S. I section B for a more detailed discussion of the low field magnetoresistance. A second difference between R_c and R_a is the amplitude of the variation caused by the phase transition. In the case of R_a , it is less than a factor of two, whereas in the case of R_c , it is several orders of magnitude. The remarkable sensitivity of the c -axis transport has been noticed previously[16], but never quantified. The third difference between R_c and R_a is their diverging behavior above 53 T. In this range, R_c increases with magnetic field and this enhancement strongly depends on the temperature. On the other hand, R_a is slightly decreasing with field and does not show any variation with temperature.

Before this work, the system was believed to reenter its low-field state above 53 T[20]. Therefore, the increase in R_c above this field was unexpected. This observation motivated us to extend our investigation of R_c to higher fields (80 T) and lower temperatures (0.44K). The data

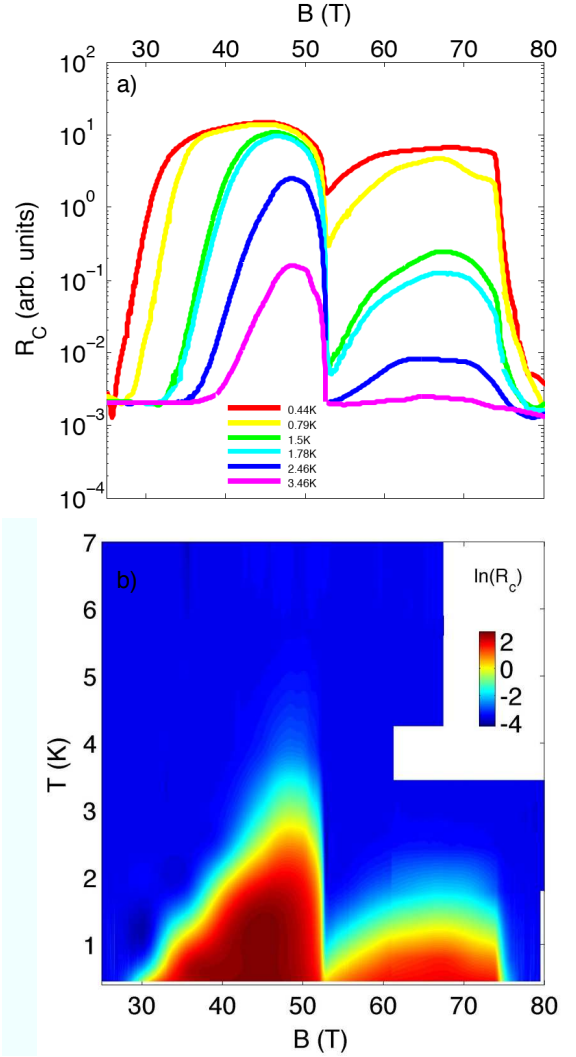


FIG. 2: a) Out-of-plane magnetoresistance in semi-logarithmic scale of kish graphite (sample K_3) up to 80 T for the lowest investigated temperatures. b) A logarithmic color map of R_c (sample K_3) in the plane of temperature and magnetic field.

are presented in Fig.2.a. As seen in the figure, above 53 T, R_c increases, saturates and then steeply drops at 75 T to recover its magnitude before the onset of transition.

Thus, R_c is enhanced by several orders of magnitude in two adjacent yet distinct field windows. The first extends between 30 T to 53 T and the second between 53 T and 75 T. In both windows, the amplitude of R_c becomes extremely temperature-dependent. Fig.2.b presents temperature and field dependence of R_c in a logarithmic color map. A new dome clearly emerges above 53 T. This corresponds to the boundaries of a new electronic phase in the (temperature, field) plane. The magnetoresistance recovers its normal amplitude above 3.5 K. This is the highest critical temperature of this new state. The two field scales 53 T and 75 T are temperature-independent

contrary to the onset of the first transition which increases with increasing temperature. The identification of an additional phase in the phase diagram of graphite beyond the quantum limit is the first result of this work.

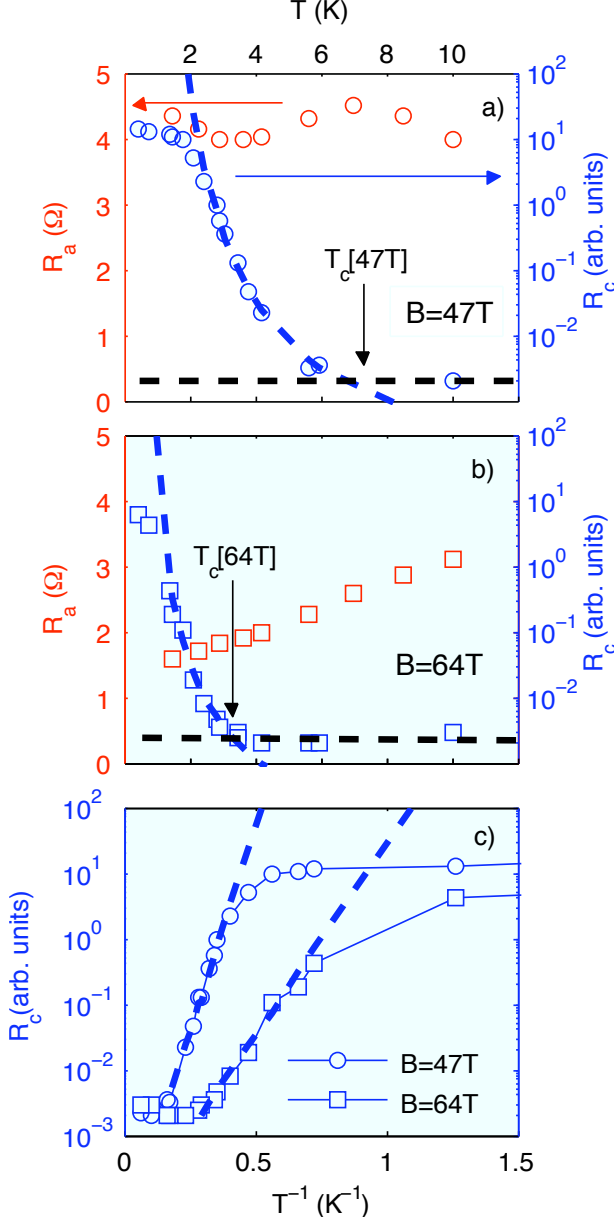


FIG. 3: Temperature dependence of the in-plane (red left axis) and out-of-plane (blue right axis in log scale) magnetoresistance of kish graphite a) for $B=47$ T (circles points) and b) for $B=64$ T (square points) for the two samples. Activated (temperature-independent) variations are represented by blue (black) dashed lines. The critical temperature of the field-induced transition is defined as the intercept of these two lines. c) R_c as a function of T^{-1} for $B=47$ T (circles) and $B=64$ T (squares). Dashed lines are Arrhenius fits to R_c .

The second result is presented in Fig.3, which compares the temperature-dependence of the in-plane and

out-of-plane resistivity in the two ordered states. In Fig.3.a) and b) (red left scale), data for R_a are presented for $B=47$ T and $B=64$ T. At each of these fields, upon cooling, the system enters deep inside one of the two ordered states (see Fig.2b). In neither case, the in-plane resistivity displays an insulating behavior in the ordered state. The temperature dependence of R_c for the same fields is presented in Fig.3.a) and b) (blue right scale). For both fields, plotting R_c as a function of T^{-1} (Fig.2.c)) reveals an Arrhenius behavior upon the entry to the ordered state. In both cases, at low temperature, resistance deviates from the Arrhenius behavior and saturates to a finite value pointing to the survival of a residual conductivity along c-axis at low temperature. We will come back to the origin of this residual c-axis conductivity in the discussion below.

By fitting the data in the activated regime to $R_c = R_{0,c} \exp(\frac{2\Delta}{k_B T})$, we found activation gaps to be $2\Delta[47T] = 2.4 meV$ and $2\Delta[64T] = 1.1 meV$. For each field, a critical temperature (T_c) for these transitions can be defined as the temperature at which the Arrhenius fit (blue dot lines on Fig.3.a) and b)) crosses the normal state resistance value (black dot lines on Fig.3.a) and b)). This yields $T_c[47T] = 7 \pm 0.5 K$ and $T_c[64T] = 3.5 \pm 0.5 K$. Comparing the experimentally-resolved activation gap with the critical temperature, we find a similar ratio for both states, $\frac{2\Delta}{k_B T_c}[47T] = 3.9$ and $\frac{2\Delta}{k_B T_c}[64T] = 3.6$. We note however a difference between the two field induced states since $\Delta[47T] \approx \frac{\Delta[64T]}{2}$.

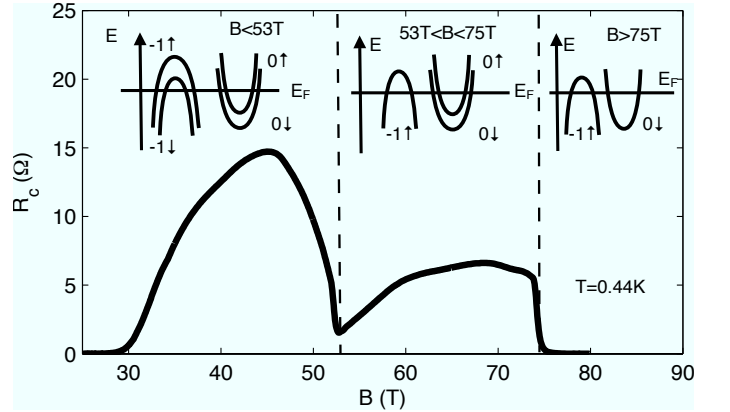


FIG. 4: Field dependence of the out-of-plane resistance R_c of kish graphite (sample K_3) at $T=0.44 K$ (black line). Sketch of occupied Landau sub-levels with increasing magnetic field. When $B > 7.5$ T, there are four occupied sub-levels. The onset of field-induced transition at 30 T, opens a gap in all these sub-levels. At $B=53$ T, one sub-level crosses the Fermi energy and the field-induced state is destroyed. For $B > 53$ T, a second field-induced state emerges opening a gap in the three last occupied sub-levels. At $B=75$ T, another sub-level becomes empty and the second field-induced state is destroyed.

These observations lead to a deep revision of our current understanding of the physics of the field-induced phase transition in graphite. Two possible configurations

have been proposed for the density wave observed below 53 T. Either two charge density waves (per valley) formed in both ($n=0, \downarrow$) and ($n=0, \uparrow$) sub-levels of electrons (and respectively in both ($n=-1, \downarrow$) and ($n=-1, \uparrow$) sub-levels of holes)[18] or one spin density wave (per valley) in each electron and hole pocket[22]. Yaguchi and Singleton[15] suggested that the two Landau sub-levels ($n=0, \uparrow$) (for the electrons) and ($n=-1, \downarrow$) (for the holes) are depopulated at 53 T and this destroys the CDW state. In this case, above this field, the two available Landau levels would be ($n=0, \downarrow$) and ($n=-1, \uparrow$). Following this picture, one should assume that the system undergoes a second density-wave instability along the c -axis at 53 T. In this case, graphite would host a cascade of density-wave transitions, a scenario reminiscent of what has been proposed in the case of the quasi-one dimensional organic conductor $(\text{Per})_2\text{Pt}(\text{mnt})_2$ [24].

However, this interpretation is challenged by the small and almost temperature-independent resistivity detected above 75 T. In the picture drawn above, when the magnetic field exceeds 53 T, only the two last spin-polarized Landau sub-levels would remain occupied. This reduces the number of possible configurations for any nesting scenario. If two sub-levels empty at 53 T and two others at 75 T, one would expect an ultimate high-field semiconducting state in contrast to what is observed experimentally. Persistence of a sizable conductivity above 75 T is not compatible with a total depopulation of all Landau sub-levels above this field.

A tempting solution would be to revise this scenario and assume that at $B=53$ T only one sub-level (instead of two) depopulates (see the sketch in Fig.4). This could be either of the two ($n=0, \uparrow$) or ($n=-1, \downarrow$) Landau sub-levels. In this case, for $B > 53$ T, there are three occupied sub-levels and a second gapped state. At $B=75$ T, the other sub-level is depopulated and the ordered state is destroyed. In this case, beyond 75 T, the ultimate electron and hole Landau sub-levels will remain full, in agreement with the finite conductivity observed by our experiment. In the SWM model, which is in very good agreement with the experimental low-field Landau spectrum [6, 25], the hole-like Fermi surface has a slightly smaller cross section than the electron-like one. Therefore, the ($n=-1, \downarrow$) sub-level is expected to become empty at a lower magnetic field than the ($n=0, \uparrow$) sub-level (as sketched in Fig.4). However, according to Takada and Goto[21], electron correlations modify the SWM spectrum at high magnetic field. Thus, the identity of the occupied sub-level between 53 T and 75 T remains an open question. This sub-level plays a crucial role in any nesting scenario, since both phase transitions occur when it is occupied and the order is destroyed as soon as it becomes empty.

Our most striking result is the observed difference between R_a and R_c . To the best of our knowledge, the field-induced state in graphite is the only case in which activated conductivity along one axis coexists with metallic conductivity perpendicular to it. This dichotomy con-

trasts with what has been reported in other density-wave systems. Bechgaard salts are a well-documented family of quasi-one dimensional conductors hosting a density-wave transition as a consequence of nesting. In the spin-density-wave of $(\text{TMTSF})_2\text{PF}_6$, all three components of resistivity display an activated behavior with a gap of similar amplitude[26]. This is not surprising, since such a transition opens a gap in the Density Of States (DOS), which affects the conductivity along all orientations. The main particularity of graphite is the presence of a quantizing magnetic field, which has already opened a cyclotron gap (as long as the chemical potential resides between two Landau levels or sub-levels). The energy distance between two sub-levels is at least the Zeeman energy, $g\mu_B B$ (where $g=2.5$ [27]), which at 53 T is as large as 8 meV. In a three-dimensional solid, the cyclotron gap does not destroy metallicity, but with the opening of the field-induced gap, which destroy the z -axis dispersion, the spectrum for bulk electrons becomes fully gapped. In this context, the in-plane metallicity of bulk electrons is hard to understand. Therefore, another source of metallicity, such as edge states, is to be considered.

The fate of a 3D electron gas system beyond the quantum limit has been the subject of several theoretical studies[8, 28, 29]. As first pointed by Halperin[8], quantum Hall effect can occur in three dimensions if the Fermi level of the bulk happens to lie inside an energy gap. A bulk density wave state along the c -axis would naturally fulfill such a condition. A theory for quantum Hall effect in graphite[28] predicted the appearance of chiral surface states[30] characterized by a ballistic in-plane longitudinal response.

It is tempting to link the in-plane metallicity observed here with such states. The order of magnitude of the observed metallicity is compatible with such an interpretation. As seen in Fig.3.b), the residual temperature-independent conductivity is $\approx 1\Omega^{-1}$ at 64 T. The thickness of our sample is $50\mu m$, approximately 150000 graphene layers. Thus, the measured in-plane corresponds to a conductivity of $0.15\sigma_0$ per graphene layer ($\sigma_0 = \frac{e^2}{h}$ is the quantum of conductance). Since for both electrons and holes, $2k_F$ is about half of the Brillouin zone, in the simplest version of the density-wave scenario, the system becomes a stack of weakly-coupled graphene bilayers. Assuming ballistic edge transport in each bilayer, one would expect a conductivity of $0.5\sigma_0$ per graphene layer, three times larger than what is found in our experiment. However, the measured intralayer and interlayer resistivities are to be treated cautiously. Only their order of magnitude remains reliable, since as a consequence of stacking defaults, in-plane and out-of-plane charge flows mix up in graphite.

Chiral edge states have been observed in the multilayer semiconductor $\text{GaAs}/\text{Al}_{0.1}\text{Ga}_{0.9}\text{As}$ by Druist *et al.*[31] who also found that these states have a finite vertical conductance. It would be natural to expect edge states in graphite to be the same. As mentioned above, a residual out-of plane conductivity survives in both ordered

states at low temperature (Fig. 3.c). A possible source of this are impurity states, known to lead to a deviation from activated behavior in conventional Density Wave systems at low temperatures[32]. We note, however, that the order of magnitude of the c -axis residual conductivity resolved here corresponds to what is expected for edge states. At zero magnetic field, the out-of-plane conductivity of graphite is much lower than in-plane conductivity (see the SI section). In the presence of edge states extending in the plane parallel to the magnetic field, one would expect that the ratio of $\frac{\rho_c}{\rho_a}$ at high magnetic field recovers its value at zero magnetic field and this is indeed the case (see the SI section C).

In summary, we investigated magnetoresistance of

graphite up to 80T and found a new electronic phase above 53 T. The high-field phase diagram of graphite is more complex than it was thought before. We also resolved an activation behavior along the c -axis coexisting with in-plane metallicity, which may be a consequence of chiral surface states. We thank J. Alicea, A. Bangura, D. K. Maude, V. Oganessian, and Y. Takada for stimulating discussions and J. Béard, A. Zitouni for technical support during the 80 T experiment. This work is supported by the Agence Nationale de Recherche as a part of SUPERFIELD project, by a grant attributed by the Ile de France regional council and by EuroMagNET II under the EU contract number 228043.

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Supplementary Information

A. Samples and the measurement

The measurements have been performed on two kinds of graphite : kish samples from Covalent Materials Corporation and Highly Oriented Pyrolytic Graphite (HOPG) samples from SPI (grade 3). The kish samples are flakes of typical length of ≈ 1 mm and thickness of ≈ 0.1 mm. The HOPG samples are cubic with a typical length of ≈ 1 mm. We present in Tab.I the typical length of the samples used in our measurements :

The resistance was measured by a standard four probe method, with an AC excitation current varying from 0.25mA to 5mA for sample K_{2,3} and 1mA for K_{1,4}. Electrical contacts to the samples were made with silver paint. The magnetoresistance measurements were performed with AC excitation at a frequency varying from 40 kHz and 60kHz using pulsed magnetic fields up to 80 T at the LNCMI-Toulouse. Both helium 4 and helium 3 cryostats were used to attained low temperatures.

TABLE I: Sample descriptions

Sample Name	Nature	Length (mm)	Type of measurement	Geometrical Factor (cm)
K_1	kish	1.7*1.31*0.05	R_a	0.006
K_2	kish	0.72*0.66*0.11	R_c	0.462
K_3	kish	1.5*0.9*0.08	R_c	1.7
K_4	kish	1.2*1.0*0.05	R_a	0.006
H_1	HOPG	1.5*1.3*1.2	R_c	0.16

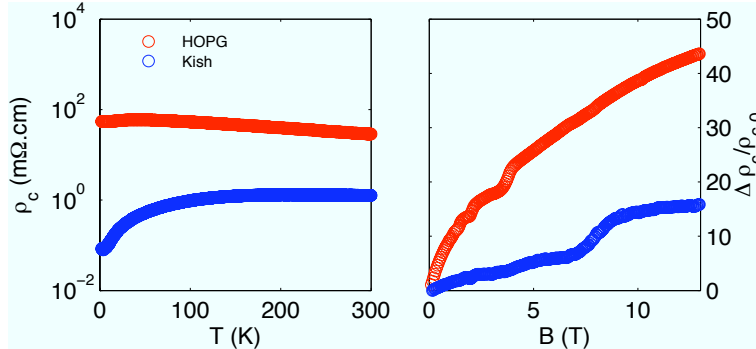


FIG. 5: a) c -axis resistivity as a function of the temperature for a HOPG sample (red points) and kish sample (blue points) b) c -axis magneto resistivity (B/c) at $T=2K$ for a HOPG sample (red points) and kish sample (blue points)

B. HOPG vs kish samples

1. c -axis temperature dependence

For both, HOPG and kish samples, the in plane resistivity at room temperature is comparable and around $70\mu\Omega.cm$. However the two systems display different c -axis behavior. Fig.5.a compares the typical temperature dependence of c -axis resistivity in HOPG and kish samples.

On one hand, the ρ_c in kish sample shows a metal-like dependence on temperature. At room temperature, $\rho_c \approx 1.2m\Omega cm$, with a typical variation of a factor two from sample to sample. Thus, the typical resistivity anisotropy ($\frac{\rho_c}{\rho_a}$) in kish samples is around 20 (five times smaller than in natural graphite [D.Z Tsang and M.S Dresseelhaus, Carbon 14,43 (1976)]).

On the other hand, ρ_c in HOPG samples increases with decreasing temperature down to 50K. It begins to show a metallic temperature dependence in lower temperatures. The typical anisotropy is much higher: $\frac{\rho_c}{\rho_a} \approx 1000$. This difference in the behavior of c -axis resistivity in the two systems has been a subject of debate. Ono *et al.* [J. Phys. Soc. Jpn 40, 498 (1976)] suggested that at high temperature HOPG is more anisotropic than kish due to a larger number of stacking faults. The metallic behavior observed below 50K has been attributed to additional mechanisms of charge transport along the c -axis. In particular, Matsubara *et al.* [PRB 41,969 (1990)] suggest that an interlayer transfer of charge can be caused by the interaction between the 2D delocalized carriers and impurity sites. In such a picture, ρ_c is naturally contaminated by ρ_a .

2. Low field c -axis magnetoresistance

Fig.5b displays the field dependence of ρ_c at $T=2K$. As reported in the literature [I.L Spain, Solid State Communications, 9, PP. 1581, 1971.], we found for both systems a sizable longitudinal magnetoresistance (still much smaller than the transverse in-plane magnetoresistance). For free electrons no longitudinal magnetoresistance is expected. Peculiar shape of the Fermi surface can lead to a non zero longitudinal magnetoresistance [23]. In the case of graphite, it has been predicted that $\frac{\Delta\rho}{\rho_0}$ should saturate to 0.12, much smaller than what is experimentally observed [23].

There are several possible origins for this discrepancy. In the case of HOPG, additional scattering mechanisms are to be considered like the conducting path mechanism. Because of the structural disorder in HOPG, a current applied along c -axis flows not only perpendicular but also parallel to the graphene layers. In such case, the c -axis

magnetoresistivity can be described by two parallel resistors involving R_c and R_a [Y.Kopelevich et al., Physics Letters A **374** (2010)].

In the case of kish, the discrepancy could be related to experimental issues. Contrary to the HOPG samples, we observe a large variation of the longitudinal magnetoresistance from sample to sample. We attributed this variations to a contamination of the large in plane magneto resistivity by a misalignment of the contacts. The real c -axis longitudinal magneto-resistance at low magnetic field in kish sample is then certainly much smaller than the observed one.

3. High-field c -axis magnetoresistance

We now turn to our high magnetic field investigation of the c -axis resistivity in HOPG samples. Fig.6 presents the field dependence of R_c in the sample H_1 at $T=1.5K$. On top of a large magnetoresistance, a double-peak structure, similar to what was observed in kish samples, emerges above 25 T. Interestingly, the boundaries of the two structures, which are respectively [31T, 53T] and [53T, 75T] match exactly the phase diagram identify in kish samples. Thus, it appears that the existence of the second field induced state is a universal property of graphitic systems.

However, we note that the quantitative analysis of R_c is more complicated in the case of HOPG than in kish graphite. In particular, the extraction of an activation gap would required a fine description of the inherent contribution of the large in-plane magnetoresistance in R_c (as discussed in B 2) and a systematic study of R_a in HOPG.

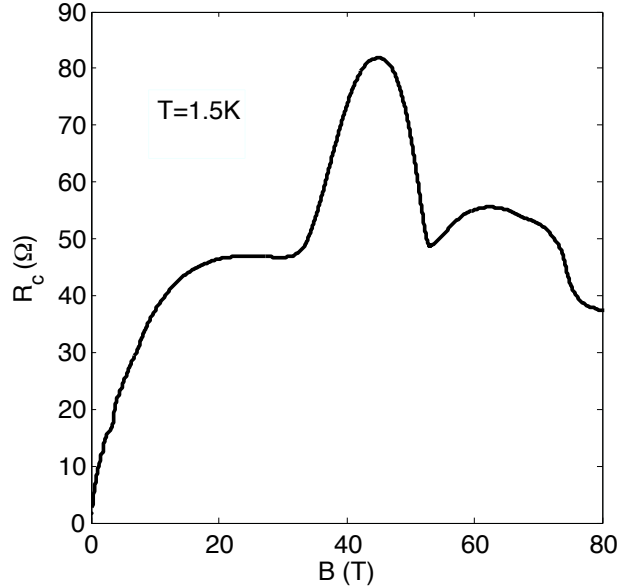


FIG. 6: a) Longitudinal resistance R_c ($B//I//c$) of the sample (H_1) as a function of the magnetic for $T=1.5K$

C. Field dependence of $\frac{\rho_c}{\rho_a}$ for kish samples

Fig.7 presents the field dependence of $\frac{\rho_c}{\rho_a}$ at $T=1.5K$ up to 70T. Due to the variation of ρ_c from sample to sample (discussed in B 1), the absolute value of this ratio should be handled with care. However, the relative variation of this ratio induced by the magnetic field is instructive. At zero field and $T=1.5K$, the typical ratio of $\frac{\rho_c}{\rho_a}$ is around 40 (comparable with the ratio of $\frac{\rho_c}{\rho_a}$ at room temperature). As the magnetic field increases, the large in-plane magnetoresistance compensates the zero-field anisotropy. At 10T, the ratio $\frac{\rho_c}{\rho_a}$ is near unity. At 25T, the field-induced state emerges. Because of the large increase in ρ_c , the ratio of $\frac{\rho_c}{\rho_a}$ drastically increases. Interestingly, in this state, the ratio becomes comparable to its zero-field value. At the reentrance field, $\frac{\rho_c}{\rho_a}$ decrease sharply followed by a re-increase due to second transition.

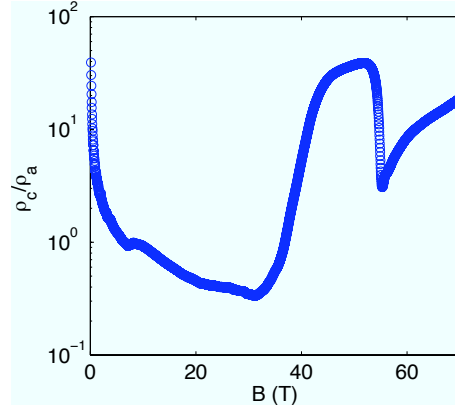


FIG. 7: Ratio of the longitudinal resistivity ρ_c (sample K₃) and the in plane resistivity ρ_a (sample K₁) as a function of the magnetic field at T=1.5K

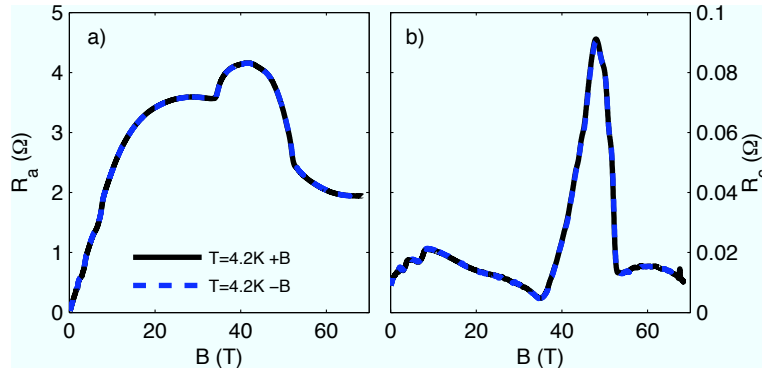


FIG. 8: a) Transversal resistance R_a of the sample K₁ b) longitudinal resistance R_c of the sample K₂ at T=4.2K for a magnetic field parallel (black line) and antiparallel (blue dot line) to the c -axis

D. Field sweeps with inverted orientations of magnetic field

On Fig.8, we present the field dependence of the in-plane and the out-of-plane resistance at T=4.2K for two opposites directions of the magnetic field parallel to the c -axis. The absence of difference between the two configurations suggest that the measured signal reflects the longitudinal component of the resistivity tensor in spite of possible contact misalignments. This is not surprising, since it is known that : $\rho_{xy} \ll \rho_{xx}$. Such equation is the result of (i) the compensated nature of the Fermi surface of graphite which gives a small Hall effect and (ii) the large mobility of the carriers which give rise to a large magnetoresistivity. The behavior of the Hall effect in the field-induced state has been so far poorly investigated. Measurements by Uji *et al.* [Physica B **246-247**, 299 (1998)] up to 30T and down to 0.5K suggest a *decrease* of the Hall effect at the transition. In other word, $\rho_{xy} \ll \rho_{xx}[\rho_{zz}]$ seems to hold also in the field induced state. An important consequence of a small Hall angle is the simplicity of the link between the resistivity and the conductivity tensors. In absence of the off-diagonal term, the link between resistivity and conductivity is simply: $\rho_{ii} = \frac{1}{\sigma_{ii}}$ where $i = x, y, z$.